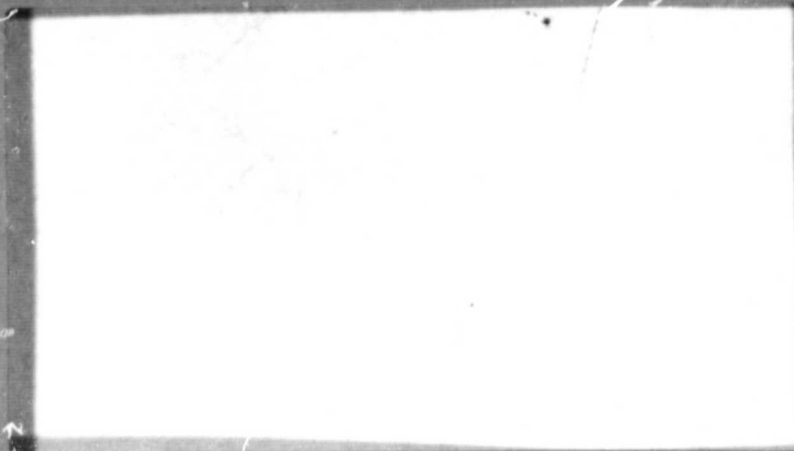


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Final Report

**DEVELOPMENT OF AN INFRARED
RADIATIVE HEATING MODEL**

SAI NO. EF79-56

April 1979

Prepared for

**Ames Research Center
National Aeronautics and Space Administration
Moffett Field, California 94035**

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I INTRODUCTION

The impact of man's activities on the atmosphere and the climatic processes are currently important questions. To help determine the effects of these perturbations, comprehensive three-dimensional numerical models have been and are being developed for predicting the atmospheric variables. Although on one hand, these models require accurate representations of the physical processes, on the other hand they also have severe requirements for computational efficiency. Maintaining this delicate balance often necessitates adapting existing techniques or developing entirely new ways of treating the processes.

Infrared radiative transfer is an important physical process in the atmosphere. Although the physics of infrared radiation is fairly well understood, the computation of accurate heating and cooling rates can be expensive; thus, approximations and simplified techniques must be used. The schemes employed in global circulation models are apparently not directly applicable to stratospheric problems. Therefore, for stratospheric numerical models new techniques or adaptations of existing techniques must be developed. This report describes the infrared radiative heating model developed for inclusion in the NASA Ames Research Center three-dimensional stratospheric numerical model.

We discuss first the background literature on both the cooling rate calculational schemes and infrared gas data. Then, we describe the line-by-line radiative transfer code which, using spectroscopic gas data, computes infrared transmittances for given absorber amounts, temperature, and pressure. Finally, we present the infrared cooling rate computer program developed for the Ames three-dimensional stratospheric model.

Several points should be emphasized here. First, the ultimate cooling rate calculation scheme has not yet been found. The cooling rate program described here represents a compromise between speed, accuracy, and developmental time. However, since we do not feel that it has the optimum time/accuracy characteristics, we suggest possible improvements. Second, the line-by-line program gives the Ames researchers the flexibility to develop their own hybrid forms of cooling rate calculations tailored to a variety of different circumstances. New and undocumented techniques, such as Wang's method, could be programmed to test the characteristics of these hybrid forms. Finally, we have included neither detailed descriptions that fully document the computer codes nor extensive examples since we view these codes as evolving. Instead, we have concentrated on presenting the rationale behind the code development and pointing to future improvements.

II BACKGROUND LITERATURE SURVEY

This section provides background data on the cooling rate calculation techniques and on laboratory data for the infrared radiative properties of atmospheric gases.

A. COOLING RATE CALCULATION TECHNIQUES

Calculations of atmospheric infrared heating and cooling rates have a long history [see Goody (1964) for a historical review]. Before large computing machines were available, graphical or tabular methods were often used (Elsasser, 1942; Yamamoto, 1952; Elsasser and Culbertson, 1960). As increasingly fast computational techniques have been developed, more extensive and detailed numerical calculations have been reported, including even extremely detailed line-by-line computations (Dickenson, 1972). The primary difficulty in performing infrared cooling calculations has been ensuring the inclusion of the important physical details without making the calculations unnecessarily burdensome and time consuming. Thus, a variety of techniques has been developed.

In computing the infrared heating/cooling rate the following choices among procedures are available:

- > Form of the radiative transfer equation:
 - Flux-differencing
 - Direct cooling rate calculation
 - Parameterization.
- > Frequency integration:
 - Line-by-line calculation
 - Band model
 - Random or Goody model
 - Malkmus model

- Exponential model
- Empirical fit
- Emissivities.
- > Path integration (inhomogeneous effects):
 - Curtis-Godsen approximation
 - Differential formulation.
- > Angle integration:
 - Diffusivity factor (exponential integral approximation)
 - Numerical integration.

A more detailed discussion of the cooling rate calculation literature is included in Appendix A. A summary of the literature is contained in Table 1.

B. LABORATORY DATA

The published data sets for the infrared radiative properties of atmospheric gases are summarized in Table 2.

TABLE 1. SUMMARY OF THE COOLING RATE CALCULATION LITERATURE

| Investigator | Gas | Band | Data Source | Computational Technique | * Accuracy (%) | Estimated 7600 Computer Time to Calculate |
|----------------------------|---|------------------|-----------------------------|--|----------------|---|
| Rogers and Walshaw (1966) | H ₂ O | rot. | Goody (1964) | Random model | 5 - 10 | 1 sec |
| | H ₂ O | 6.3 μ | Burch et al. (1962) | Random model | | |
| | H ₂ O | window | Bignelli et al. (1963) | Constant K | | |
| | CO ₂ | 15 μ | Houghton and Rodgers (1966) | Random model | | |
| | O ₃ | 9.6 μ | Walshaw and Rodgers (1963) | Random model | | |
| Ramanathan (1976) | H ₂ O | rot. + 6.3 μ | Staley and Jurica (1970) | Emissivity | -- | -- |
| | H ₂ O | window | Bignelli (1970) | | | |
| | CO ₂ | 15 μ | Dickenson (1972) | Band absorp- tance | | |
| | O ₃ | 9.6 μ | Aida (1975) | Band absorp- tance | | |
| | | | | | | |
| Rodgers (1967) | O ₃ | 9.6 μ | Walshaw and Rodgers (1963) | Random model | -- | -- |
| Dickenson (1972) | CO ₂ | 15 μ | Line data | Line-by-line code (escape functions) | 1 | -- |
| | | | | | | |
| Cess (1974) | O ₃ | 9.6 μ | Line Data | | | |
| | H ₂ O | all | Hottel (1954) | Emissivity | -- | -- |
| Ellingson and Gille (1978) | H ₂ O | all | Benedict and Calfee (1967) | Random model | 3 | -- |
| | | 15 μ | Goody (1964) | Random model | | |
| | CO ₂ | 15 μ | Drayson and Young (1967) | Random model | | |
| | O ₃ | -- | McClatchey (1972) | Random model | | |
| | (CH ₄ , N ₂ O) | | | | | |

TABLE 1 (Continued)

| Investigator | Gas | Band | Data Source | Computational Technique | Accuracy* (%) | Estimated 7600 Computer Time to Calculate |
|----------------------------------|------------------|----------------|--|--|---------------|---|
| Hunt and Mattingly (1976) | H ₂ O | all | AFCRL tape McClatchey et al. (1973) [†] | Random model | -- | -- |
| | CO ₂ | 15 μ | McClatchey et al. (1973) | Random model | | |
| | O ₃ | 9.6 μ | McClatchey et al. (1973) | Random model | | |
| Ackerman, Liou, and Leovy (1976) | H ₂ O | all | Rodgers and Walshaw (1966) | Sum of exponentials | -- | -- |
| | CO ₂ | 15 μ | Rodgers and Walshaw (1966) | Sum of exponentials | | |
| | Aero-sols | 8-12 μ | Calculated | Sum of exponentials | | |
| Kuo (1977) | H ₂ O | 6.3 μ rot. | Goody (1964) | Exponential band model | -- | -- |
| | CO ₂ | 15 μ | Benedict and Calfee (1967) Line data | Exponential band model | | |
| | O ₃ | 9.6 μ | Walshaw (1957) | Exponential band model | | |
| Fels and Schwarzkopf (1979) | CO ₂ | 15 μ | Drayson (1967) line data | Line-by-line to parameterized (matrix) | -- | -- |
| Chou and Arking (1978a, 1978b) | H ₂ O | all | McClatchey et al. (1973) line data | Line-by-line to parameterized | 10 | -- |
| | CO ₂ | 15 μ | McClatchey et al. (1973) | Line-by-line to parameterized | | |

TABLE 1 (Concluded)

| Investigator | Gas | Band | Data Source | Computational Technique | * Accuracy (%) | Estimated 7600 Computer Time to Calculate |
|----------------------|------------------|-------------------------|--------------------------------------|----------------------------------|----------------|---|
| Wu (1976) | H ₂ O | 6.3 μ , rot. window | Benedict and Calfee (1976) line data | Random model | -- | -- |
| | H ₂ O | 15 μ | Lee (1973) | Formula for dimer | -- | -- |
| | CO ₂ | 15 μ | Line data | Transmission function | -- | -- |
| Coakely (1978) | CO ₂ | 15 μ | Burch et al. (1962) | Band model | -- | -- |
| Wang and Laci (1978) | O ₃ | 9.6 μ | McClatchey (1973) Line-by-line | Malkmus model to exponential sum | 2 | -- |
| | H ₂ O | all | McClatchey (1973) | Malkmus model to exponential sum | -- | -- |
| | CO ₂ | 15 μ | McClatchey (1973) | Malkmus model to exponential sum | -- | -- |

* "Accuracy" as expressed in this column is intended as a general term and not as the result of one specific calculation.

+ AFCRL = Air Force Cambridge Research Laboratory.

TABLE 2. BRIEF SUMMARY OF AVAILABLE DATA

| Investigator | Gas | Band | Method | Range | | | Accuracy (%) |
|--|------------------|-------------|--|-------------------|---------------------|------------------------------|-----------------|
| | | | | Pressure (atm) | Temperature (°K) | Absorber Amount (ppcm) | |
| Burch et al. (1962) | H ₂ O | all | Long path spectrometer | 0.01 - 1 | ~296 | 0.001 - 0.1 | ~5 |
| | CO ₂ | 15 μ m | Long path spectrometer | 0.01 - 1 | ~296 | 0.001 - 0.1 | ~5 |
| Howard et al. (1955) | H ₂ O | all | Long path spectrometer | 0.1 - 1 | ~296 | 0.01 - 0.1 | ~5 |
| | CO ₂ | 15 μ m | Long path spectrometer | 0.1 - 1 | ~296 | 0.01 - 0.1 | ~5 |
| Palmer (1960) | H ₂ O | window | Long path spectrometer | 0.01 - 1 | ~296 | 0.001 - 0.1 | 3 |
| Roberts, Biberman, and Selby (1976) | H ₂ O | window | Transmission measurements | 1 | ~296 | -- | -- |
| Bignell (1970) | H ₂ O | window | Long path spectrometer | 1 | 33 - 45 (°C) | 0.1 - 1 | -- |
| Stauffer and Walsh (1966) | H ₂ O | window | Long path spectrometer | 0.1 - 1 | 296 | 0.001 - 0.1 | ±2 |
| Ferriso, Ludwig, and Thomson (1966) | H ₂ O | all | Statistical model fit to data | ~1 | 300 - 3000 | -- | ±20 |
| McClatchey et al. (1973) | H ₂ O | all | Compilation of line data (spectroscopic) | | STP | | -- |
| | CO ₂ | 15 μ m | Compilation of line data (spectroscopic) | | STP | | -- |
| | O ₃ | 9.6 μ m | Compilation of line data (spectroscopic) | | STP | | -- |

TABLE 2 (Concluded)

| Investigator | Gas | Band | Method | Range | | | Accuracy (%) |
|----------------------------|------------------|-------------|--------------------------------|-------------------|---------------------|------------------------------|-----------------|
| | | | | Pressure (atm) | Temperature (°K) | Absorber Amount (ppcm) | |
| Benedict and Calfee (1967) | H ₂ O | 6.3 μ m | Line data | | STP | | -- |
| | CO ₂ | 15 μ m | Line data | | STP | | -- |
| Aida (1975) | O ₃ | 9.6 μ m | Quantum mechanics calculations | | STP | | -- |

III INFRARED TRANSMISSIONS OF ATMOSPHERIC GASES

The "first principles" approach to infrared transmission of atmospheric gases is to first solve the quantum mechanics equations for the states of the particular molecule. One can then compute the absorption coefficients from the transition probabilities and energy levels. In practice, this approach is rarely used because most molecules are too complicated to yield a solution to the wave equations. What can be done is to use either experimentally determined or computed line position, line strength, and half width data to compute the absorption coefficient of the gas. This procedure of computing the spectral absorption coefficient of individual lines is called the line-by-line calculation procedure. Only recently, with the availability of sufficient data on atmospheric gases and with high-speed computers, has this technique become practical. This section of the report describes the line-by-line transmission computer program, which allows the user to develop a variety of transmission parameterizations for specific uses.

A. LINE-BY-LINE CALCULATIONS

The physics of infrared radiation and molecular structure is described by Herzberg (1945), Goody (1964), and others. Basically, an energy transition from State 1 to State 2 gives rise to an absorption (emission) coefficient with a particular spectral shape depending upon the dominant physical mechanisms. The three most common broadening mechanisms are natural, pressure (or Lorentz), and thermal (or Doppler) broadening.

Natural broadening is actually an effect of the fundamental uncertainty as to the location of the line. The absorption coefficient for a line for natural broadening is:

$$K_N = \frac{1}{\pi} \frac{S\alpha_N}{(n - n_0)^2 + \alpha_N^2} \quad , \quad (1)$$

where K_N is the absorption coefficient or cross section in $\text{cm}^2/\text{molecule}$, S is the line strength, α_N is the natural broadening half width, and n_0 is the wavenumber location of the line center.

Pressure (or Lorentz) broadening is an effect of the collisions between molecules. This effect is proportional to the pressure, hence the name pressure broadening. The absorption coefficient for a line for Lorentz broadening is

$$K_L = \frac{1}{\pi} \frac{S \alpha_L}{(n - n_0)^2 + \alpha_L^2} \quad (2)$$

where α_L is the pressure broadened half width (proportional to the total pressure).

Thermal (or Doppler) broadening is caused by the thermal motion of the molecule and is proportional to the temperature. The absorption coefficient for a line for Doppler broadening is

$$K_D = \frac{S}{\alpha_D} \exp \left[- \frac{(n - n_0)^2}{\alpha_D^2} \right] \quad (3)$$

where α_D is the Doppler broadening halfwidth.

In the lower troposphere the pressure broadening dominates and the lines are essentially Lorentzian. However, in the upper stratosphere the pressure is low enough that Doppler broadening becomes important; it dominates at the very highest altitudes. Thus, there is a region where both Lorentz and Doppler broadening are important. The combination of Lorentz and Doppler broadening, known as the Voigt profile, can be written

$$K_V = \frac{1}{\alpha} \int_{-\infty}^{\infty} K_L(n') \cdot K_D(n - n') \, dn' \quad (4)$$

The Voigt profile expression is thus not analytic and requires the evaluation of the integral in Eq. (4).

To evaluate the absorption coefficient of the gas at a particular wave-number it is necessary to sum the absorption coefficients at that wavenumber from all the lines, i.e.,

$$K = \sum_{j=1}^M K_j^n (n - n_{0,j}) \quad (5)$$

where $n_{0,j}$ is the location of the center of the j -th absorption line. Although the procedure is straightforward, in the case of an infrared vibrational-rotational band there can be as many as 100,000 lines.

In the next section, we describe the computer program that evaluates Eqs. (1) through (5) for O_3 , H_2O , and CO_2 for atmospheric conditions.

B. OUTLINE OF THE COMPUTER CODE

The flow diagram for the computer code is shown in Figure 1. The main program, TRANS, contains the line data read from the AFCRL tape. The VTABLE subroutine calculates a table of Voigt profile values by solving the integral of Eq. (4) in three different ways. The transmission over a given spectral interval is defined as

$$T_{\Delta n}(U, T, P) \equiv \frac{1}{\Delta n} \int_{\Delta n} \exp[-KU] \, dn \quad (6)$$

where U is the absorber amount, T is the temperature and P is the pressure. The transmission is calculated for a Voigt profile by subroutine VOIGT and for a Lorentz profile by subroutine LORENTZ.

The equivalent width, the quantity most often reported in the literature, is defined as

$$A(U, T, P) \equiv \int_{-\infty}^{\infty} [1 - \exp(-KU)] \, dn \quad (7)$$

The equivalent width is then plotted out by routine EQWIND.

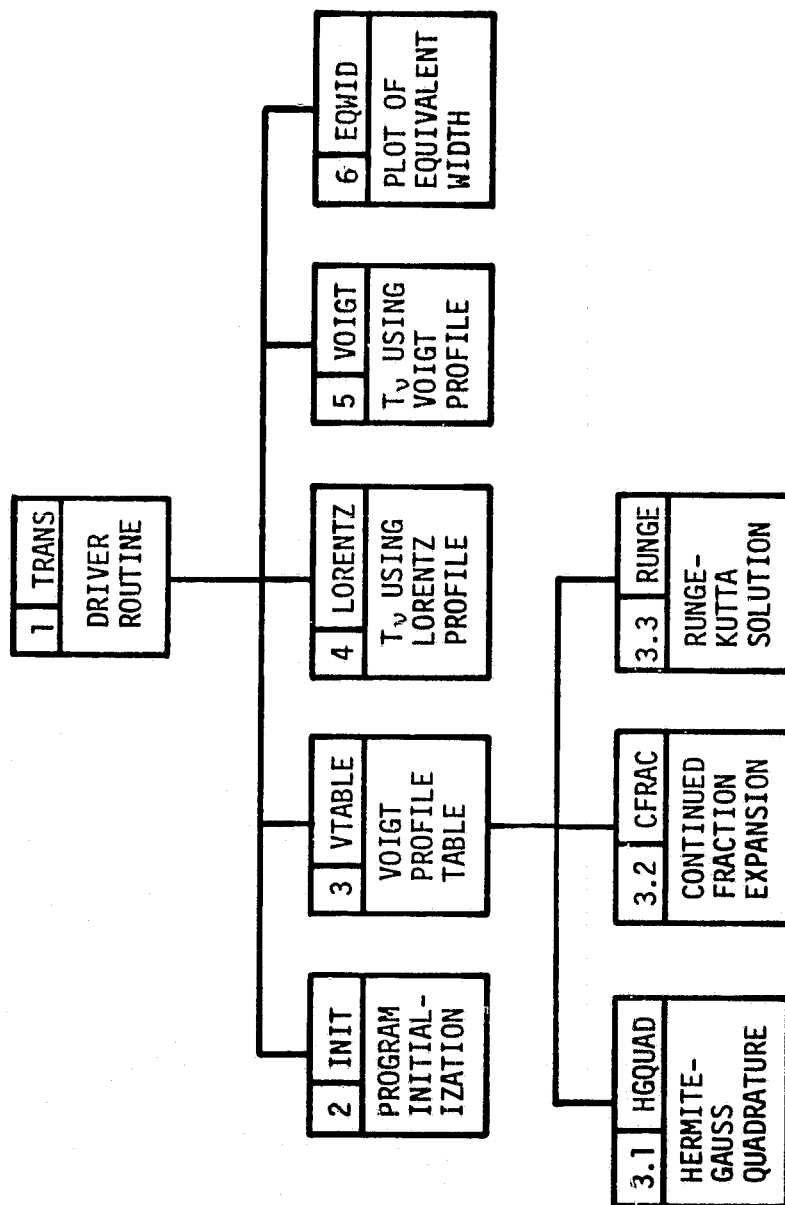


FIGURE 1. FLOW DIAGRAM FOR THE LINE-BY-LINE CALCULATION PROCEDURE

The program has been tested by comparing the results for the $15\text{ }\mu\text{m CO}_2$ band with the measurements of Burch, et al. (1962). The results are generally good but a detailed analysis has not been performed.

IV ATMOSPHERIC COOLING RATE CALCULATIONS

This chapter describes the creation of the computer program to compute atmospheric cooling rates. The underlying equations are presented followed by an outline of the computer code and a discussion of the testing results.

A. FORMULATION.

The starting point for the calculation of infrared radiative transfer is the radiative transfer equation for a plane parallel atmosphere with no scattering:

$$\mu \frac{dI_v}{dz} = \rho K_v (B_v - I_v) \quad (8)$$

where μ is the cosine of the zenith angle, I_v is the radiant intensity at frequency v , K_v is the absorption coefficient, B_v is the Black Body intensity, and z is the vertical coordinate. Integrating the equation to write the intensity in the positive z direction yields

$$I_v^+(z, \mu) = B_v(0) T_v(z, 0) + \int_0^z B_v(z') \cdot \rho K_v T_v(z, z') \frac{dz'}{\mu} \quad (9)$$

where

$$T_v(z_1, z_2) \equiv \exp \left[\frac{-\rho K_v (z_1 - z_2)}{\mu} \right]$$

If we now integrate Eq. (2) over the hemisphere, the radiant flux, F , normal to z can be written as

$$\begin{aligned}
F_v^+(z) &= \int_0^{2\pi} \int_0^1 I_v^+(z, \mu) \mu d\mu d\phi \\
&= 2\pi B_v(0) T_v^f(z, 0) \\
&\quad + 2\pi \int_0^z B_v(z') K_v T_v^f(z, z') dz' \quad (10)
\end{aligned}$$

where now

$$\begin{aligned}
T_v^f(z_1, z_2) &\equiv \int_0^1 \exp\left[\frac{-K_v \rho (z_1 - z_2)}{\mu}\right] \mu d\mu \\
&= 2 E_3[K_v(z_1 - z_2)] \quad .
\end{aligned}$$

The flux can also be expressed as

$$\begin{aligned}
F_v^+(z) &= 2\pi B_v(0) \cdot T_v^f(z, 0) \\
&\quad + 2\pi \int_0^z B_v(z') \frac{dT_v^f}{dz'}(z, z') dz' \quad . \quad (11)
\end{aligned}$$

The radiative cooling rate is defined

$$\rho c_p \left(\frac{\partial T}{\partial t} \right)_{\text{rad}} \equiv - \frac{\partial F(z)}{\partial z} \quad . \quad (12)$$

There are generally two different ways of computing Eq. (12). The first is simply

$$\frac{\partial F}{\partial z}(z) \equiv \frac{F(z_{j+1}) - F(z_{j-1})}{2\Delta z_j} \quad (13)$$

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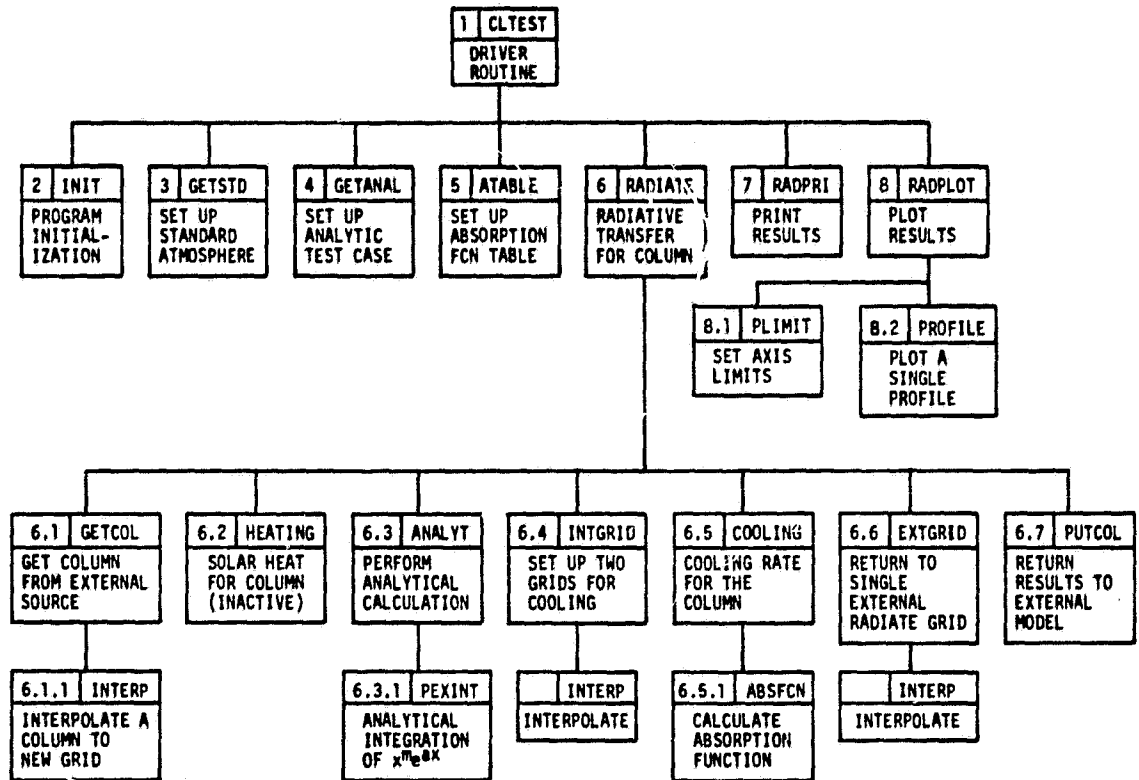


FIGURE 2. FLOW DIAGRAM FOR THE LINE-BY-LINE CALCULATION PROCEDURE

C. DISCUSSION

Initial testing revealed that a straightforward, accurate evaluation of Eq. (14) produced cooling rates with a computational speed of roughly two orders of magnitude too slow for inclusion in the Three-Dimensional Stratospheric Model. Thus, considerable acceleration of the code is necessary. We have tried the following modifications with the listed success or problem as a result.

| <u>Modification</u> | <u>Result</u> |
|--|-------------------------------------|
| Tabular values of the absorption | Approximate 50% reduction in time |
| Reduction in levels from 40 | Severe loss of accuracy |
| Two-grid system--one for cool-to-space and one for exchange integral | Marginal improvement |
| Variation in z, z' spacing in Eq. (14) | Approximate factor of 2 improvement |

As the above tabulation shows, we have been able to bring the program to within one order of magnitude of the required speed. However, meeting the requirements of the NASA Ames Three-Dimensional Numerical Model will require further testing. We discuss the possible improvements in the next section.

V RECOMMENDATIONS FOR FURTHER WORK

As discussed previously, we have developed programs that give the NASA Ames researchers a great deal of flexibility in calculating infrared cooling rates. Unfortunately, the straightforward evaluation of the cooling rate equation requires substantially more time than is desired by the Three-Dimensional Stratospheric Model. Although we have succeeded in accelerating the code by a considerable amount, it is not certain that this approach will yield the desired speed/accuracy criteria. Possibly, no scheme can give the desired results at present. On this pessimistic note we recommend that the following work be undertaken:

- > Continue attempts to accelerate the code. It may be possible to eliminate parts of the exchange integral evaluation.
- > Explore new techniques. The only technique that is substantially different is that of Wang (Wang and Lacis, 1979).
- > Compute the cooling for all three gases and account for overlap.
- > Account for nonhomogeneous paths.

We feel that the first recommendation is the most important, for if this approach is successful, the second effort would be unnecessary. The third and fourth recommendations are merely a matter of implementing known techniques and require no new development.

APPENDIX A
SUMMARY OF MEETINGS ON INFRARED
HEATING/COOLING RATE CALCULATION

APPENDIX
SUMMARY OF MEETINGS ON
INFRARED HEATING/COOLING
RATE CALCULATION

Appendix

SUMMARY OF MEETINGS ON INFRARED

HEATING/COOLING RATE CALCULATION

at

NASA Ames Research Center
Moffett Field. Ca.

5-21 July 1978

Participants:

J. B. Pollack
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Prepared by
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28 July 1978

The following is a summary of the author's notes from the set of meetings on Infrared Heating Rate Calculations at NASA Ames Research Center July 5-28, 1978. Also included are corrections and comments supplied by the meeting participants. The meetings were an informal discussion about the techniques that are currently being used and or being developed to compute infrared radiative transfer. This report is designed to document the opinions expressed at the meetings.

There were 7 meetings held over a two and one-half week period. The first meeting was an introductory one, the next four dealt with specific gaseous absorbers, the sixth with numerical methods and the last was a potpourri. This report will give a synopsis of each meeting and then a summary and recommended course of action as of 28 July 1978 for developing a radiative transfer model for inclusion in the NASA Ames three-dimensional numerical model.

Meeting 1: July 5, 1978

Some of the unique requirements for the radiation model for the three-dimensional stratospheric numerical model were discussed. They are the following:

- > Vertical resolution from the surface to 90 km. This makes Doppler effects important and possible non LTE effects.
- > Severe computational efficiency requirements $\sim 10^{-3}$ seconds per vertical column.

Other uses of infrared radiation heating model (or variations of it) are the following:

- > Two dimensional climate model.
- > Photochemical stratospheric model.
- > Time marching one-dimensional climate model.

Previous Ames radiation models were described. Emphasis has been placed on comparison to laboratory measurements. Some of the concerns revolve around the problems of Doppler broadening, inhomogeneous temperature and pressure effects, minor gaseous constituents and aerosol infrared effects. A decision was made to devote a session's discussion to the three major gases of importance (CO_2 , H_2O , O_3), a session on minor constituents and one on numerical problems.

Meeting 2: July 7, 1978

The topic of discussion was the $15\ \mu\text{m}$ CO_2 band. The discussion proceeded from paper to paper in a somewhat arbitrary fashion.

1. Rodgers and Walshaw, 1966.

- > Goody random model with quantum mechanical values for the model parameters.
- > Problems brought up to JAC
 - 1) Cooling rates do not agree with Dickinson's line by line calculation.
 - 2) The Goody random model becomes saturated at present conditions.
 - 3) In general lab data is only applicable to the troposphere so that the stratosphere must be modeled. Then the problem is how to match the two regions. Line by line calculations are OK for the stratosphere but the overlapping in the troposphere is difficult to account for. In the troposphere one should compare to laboratory equivalent widths.

2. Dickinson, 1973.

- > State-of-the-art calculation. Worked in the nonoverlapping regime, summed up equivalent widths, computed escape-to-space functions for Voigt profile. Included the hotbands, isotopic bands and temperature dependence. Dickinson reduced the thousand or so lines in each of the vibration-rotation bands of CO_2 to 10 equivalent lines. these equivalent lines are listed in his paper on the thermal structure of the atmosphere of Venus. Also the contribution of NLTE effects in Dickinson, 1973 are listed for each band.
- > Non LTE effects were 20 percent @ 85 km for $^{12}\text{C}\ ^{16}\text{O}_2$.
- > Standard for comparison.

3. Kuo, 1977.

- > Developed a generalized absorption coefficient. Fit a linear curve to the fall off with frequency (similar to the exponential band model of Edwards and Menard).
- > Kuo's method appears to have general applicability. Extension of his method, however, requires that the temperature dependence be evaluated for the distribution of absorption coefficients.

4. Fels, unpublished (Davis conference).

- > Uses a tabulated line by line calculation (Drayson).
Fels produces for present atmospheric conditions a matrix of transmissions between preassigned atmospheric layers. He also provides derivatives of the matrix elements for $\pm 20^\circ$ K excursions from the standard temperature profiles. His method is not a matrix analogue of Newtonian cooling. An example of a matrix analogue to Newtonian cooling is presented in a paper which appeared in JAS, 34, 1402 (1977). Finally, since Fels's calculations are line-by-line calculations it's not clear whether the transmissions he produces in the troposphere match laboratory measurements.
- > Problems:
 - 1) CO_2 amount can't change.
 - 2) extension to troposphere.

5. Stull, Wyatt, Plass, 1962.

- > Quasi Random band model.
- > Quantum mechanical temperature effects.
- > Used in previous Ames radiation models.

6. Coakely, unpublished (See p. 37).

- > Assumes strong Lorentz lines (good up to 50 km).
- > Uses temperature dependence.
- > Accounts for hot and isotopic bands. Uses overlap procedure of Goody and Belton.
- > Comparison to Dickinson ~ 10 percent low.

7. Ramanathan, 1976.

- > Uses Cess' band model.**
- > Includes only the P and R but not the Q branch of the band.**
- > Accounts for Doppler broadening following Cess (1978).**
Considers pure Lorentz then pure Doppler and uses the largest cooling rate.

Meeting 3: July 10, 1978

The meeting was devoted to a discussion of the water vapor effects.

A. H₂O continuum

1. Roberts, et al., 1976
 - > analysis of transmission data
 - > includes temperature effects
2. Coffey, 1977, atmospheric measurements
3. Stephens, 1976, calculations of IR cooling
4. Burch, et al., unpublished laboratory data

B. Pure rotational band

1. Stauffer and Walsh, 1966, 14-20 μ m data
2. Palmer, 1962 data
 - used in previous Ames radiation models
3. Rodgers and Walshaw, 1966
 - > Not tested against lab data (JAC tested it against Mottel's data, result poor)
 - > Should be tested against Palmer's data
4. Jurica, 1970
 - > Used Wyatt, Stull and Plass's results to compute a temperature dependent emissivity
5. Coakely, unpublished
 - > Used Walshaw and Rodgers to compute a temperature dependent emissivity
6. Ramanathan, 1976
 - > Used emissivities published by Staley and Jurica, 1970

C. 6.3 μ m band

1. Benedict et al., unpublished
 - > Quantum mechanical calculations
2. Burch et al., 1962
 - > lab data
3. Ferriso, Ludwig and Thomson, 1966
 - > lab data for higher temperatures from 1-22 μ m

Meeting 4: July 12, 1978

This meeting discussed the 9.6 μm band of ozone.

1. McClathey, et al., 1972
 - > tabulates absorption coefficient
 - > used in previous Ames models
 - > claims the pressure is less effective than gas amount due to overlapping of lines (O_3 lines are very close)
2. Aida, 1975
 - > theoretical calculation
 - > covers the non-overlapping line limit to full overlap. No Doppler effects considered. Temperature dependence found to be weak.
3. Rodgers, 1969
 - > Malkmus model fit to Walshaw's data
 - > reduces correctly to non-overlapping limit (good for small amount, small pressure)
4. Ramanathan, 1976
 - > uses Cess' band model again
 - > makes point of neglect of 9.4 μm band. However, JAC stated both Rodgers and Ramanathan's results were essentially identical.
 - > accounted for Doppler effects in the same way as for CO_2
5. Dickinson, 1973
 - > Ramanathan showed agreement to 10 percent
 - > standard to compare against for upper levels (Aida for lower)

Meeting 5: July 14, 1978

This meeting reviewed and clarified several points then discussed methane, nitrous oxide and aerosols.

1. Ramanathan's models for CO_2 and O_3
For CO_2 JAC maintained that Ramanathan did not include enough parts of the band. However, his results when compared to Dickinson are 10 percent. For ozone Ramanathan's model is OK but so is Rodgers. In general the model Ramanathan is using asymptotates correctly to the weak and strong line limit but not to the strong line overlap. This should make his results in the troposphere suspect. However, for ozone his results are 10 percent of Aida's. There was a brief discussion on inhomogeneous path problems.
2. N_2O and Methane
 - > Burch et al - data
 - > Green et al - 1964 data
3. Aerosols
 - > Wang, Domoto, 1974 - calculations
 - > Successive orders of scattering
 - > Modification of optical depth

Meeting 6: July 17, 1978

This meeting was devoted to numerical techniques.

1. Angle Integration

- > compare diffusivity factor to more accurate integration

2. Frequency Integration

- > Lowest level is 5 spectral intervals (CO_2 , O_3 , 3- H_2O)
- > JAC accounts for overlap by assuming a water vapor atmosphere then perturbs it. He uses 20 spectral intervals, 50 layers with an accurate integration. This requires $\sim 20,000$ transmission evaluations and 1-2 secs 7600 CPU time. He felt 5 spectral intervals (emissivities) can be made to be 1 percent accurate on the flux.

3. Path integral

- > Can use flux divergences for cooling rate calculations if use differential form of the equation (i.e., an equation for flux differences where $\int B dT$ becomes $\int B (T_{+1/2} - T_{-1/2})$). JAC uses essentially a Romberg integration which is very accurate (but slow). Hence the number of layers necessary is somewhat irrelevant. Normally in GCM's they use 12 layers and a trapezoidal integration. The number of transmission evaluations goes as the number of layers squared. Coakely's method is roughly 100 times as long. His estimate of the NCAR GCM's code ~ 10 percent accuracy.
- > JAC felt that in the stratosphere it is much better to evaluate cooling rates directly at the center of the layer rather than by evaluating the net flux at the top and bottom of the layer and taking the difference. Round off problems seriously compromise the latter approach.
- > For inhomogeneous paths, JBP felt that a total differential method is preferable to Curtis-Godson. If $T_r = f(T, P, U)$ goes as $(T/T_0)^n (P/P_0)^m U^e$ then he defines $\bar{K} = K_0 [P^n T^m du]^e$.

JAC provided the following rebuttal:

Comment on problem of inhomogeneous paths. For a homogeneous path the mean transmission is given by

$$T = \frac{1}{\Delta v} \int_{\Delta v} dv \exp[-\sum_i S_i g_i(v) U]$$

and presumably it can be written as

$$T = f(W)$$

where

$$W = (T/T_0)^n (P/P_0)^m U^\epsilon$$

For an inhomogeneous path

$$T = \frac{1}{\Delta v} \int_{\Delta v} dv \exp [-\sum_i S_i g_i(v) dU] .$$

Because of differences in line profiles between homogeneous and inhomogeneous paths, it need not follow that

$$T = f(W) \text{ where}$$

$$W = [\int (P/P_0)^{n/\epsilon} (T/T_0)^{m/\epsilon} dU]^\epsilon .$$

Stick with simple Curtis-Godson approximation when possible.

Problem of Doppler effects. Possibilities are the following:

- > Cess's method
- > line by line
- > Break up of Kuo's coefficient into strong line regime and Doppler regime

Meeting 7: July 21, 1978

The last meeting discussed several general topics.

1. NCAR GCM model

- > H₂O - Sasamori
- > Continuum - Roberts
- > CO₂ - Coakely
- O₃ - Rodgers

The intent is to obtain accuracies of 1 percent on flux and 10 percent on cooling. There is some question if this is achieved.

2. Coakely's CO₂ model

- > good up to 45 km, above that the results are a factor to two compared to Dickinson.
- > Doppler effects should be applied separately to each band (similar to Ramanathan)
- > There is a possibility of fitting a linear curve to the equivalent widths of Dickinson. This would allow a uniform modification instead of each band separately.
- > In the 15 μ m region CO₂ bands of nonoverlapping doppler lines seem to have an equivalent width which is proportional to $\ln(W)$ where W is the optical pathlength of CO₂.

SUMMARY AND RECOMMENDATIONS FOR DEVELOPING AN INFRARED RADIATIVE TRANSFER MODEL (RWB)

1. CO_2
 - > Dickinson, 1973 standard of comparison for upper atmosphere
 - > Coakely's model corrects the deficiencies of Ramanathan's model
2. H_2O Continuum
 - > Roberts et al. latest model
3. H_2O rotational and $5.6 \mu\text{m}$
 - > Temperature dependent emissivities, Coakely and Jurica seem promising
4. O_3
 - > Either Ramanathan or Rodger's model fits Aida calculations
 - > Ramanathan's results fit Dickinson's
5. Numerical methods
 - > Difficulty is the exchange integral evaluation (time consuming)
 - > unresolved question of how to evaluate Doppler effects

RECOMMENDATIONS FOR INFRARED CALCULATIONS FOR 3-D MODEL (as of 28 July 1978)

1. CO_2
 - > Coakely's model
 - > Doppler effects as Ramanathan
2. H_2O
 - > Either Coakely's or Jurica's emissivities
3. O_3
 - > Ramanathan's model
4. Other possibilities
 - a. Fels' method
 - b. Wang and Hansen's method
5. Problems
 - a. Doppler effects ad hoc
 - b. Probably will require more computer time than optimum
(1 millisec/column)

COAKELY'S CO₂ MODEL

The model for the equivalent width of the 15 μm CO₂ band is derived by following the procedures used to compute the equivalent widths for the vibration rotation bands of rigid rotor molecules (Cess and Tiwari). When lines do not overlap, they absorb independently of each other and the equivalent width of the band is obtained by summing the equivalent widths of the lines. When the lines overlap, the equivalent width is obtained by allowing for overlap as described by Goody and Belton (1967).

The equivalent width of a strong pressure-broadened absorption line is given by

$$A_J = 2 \sqrt{S_J \gamma_J U} \quad (A.1)$$

Where U is the absorber amount; γ is the Lorentz half-width and S_J is the line strength. If lines are non-overlapping, the equivalent width of a vibration-rotation band like those of CO₂ in the 15 μm region would be given by

$$A_0 = \sum_J A_J^P + A_J^Q + A_J^R \quad (A.2)$$

where P, Q, and R represent the three branches of the band.

In the P and R branches the strengths of the lines are approximately given by

$$S_J^{P,R} = \propto \frac{J}{2} \beta B e^{-\beta B J^2} \quad (A.3)$$

while in the Q branch they are given by

$$S_J^Q = \propto J \beta B e^{-\beta B J^2} \quad (A.4)$$

where

$$\alpha = \sum_J S_J^P + S_J^Q + S_J^R \quad (A.5)$$

is the band strength; B is the rotation constant (in cm^{-1} , for CO_2 $B = 0.39 \text{ cm}^{-1}$) and $\beta = 1.44/T$ (T in $^\circ\text{K}$).

If we insert (A.3) and (A.4) into (A.1) and then insert the result into (A.2), we find that the equivalent width for the band is given by

$$\begin{aligned} A_o &= \left[2 \int_0^\infty dJ \left(\frac{J}{2} \beta B \right)^{1/2} e^{-\frac{\beta B J^2}{2}} + \int dJ (J \beta B)^{1/2} e^{-\frac{\beta B J^2}{2}} \right] 2(\alpha \gamma U)^{1/2} \\ &= 2 \left(1 + 1/\sqrt{2} \right) \left(\frac{2}{\beta B} \right)^{1/4} \Gamma(3/4) (\alpha \gamma U)^{1/2} \quad (A.6) \end{aligned}$$

where we have assumed that all lines have the same half width, γ .

The above expression applies when all lines are present, i.e., J in (A.2) is allowed any integer value. Although all lines are present in some of the hot bands and isotopic bands in the 15 μ region, in others, like the fundamental, every other line is missing. To obtain the equivalent width when every other line is missing, we restrict the sum in (A.2) to even J only. With this modification we obtain an expression which is identical to (A.6) but with B replaced by $4B$. Thus, when every other line is missing, the equivalent width is given by

$$A_o = \frac{2}{\sqrt{2}} \left(1 + \frac{1}{\sqrt{2}} \right) \left(\frac{2}{\beta B} \right)^{1/4} \Gamma(3/4) (\alpha \gamma U)^{1/2}$$

We notice that in the above expressions, if $\sim 2(T_0/T)^{1/2}$, as it would be for pressure broadened lines having the same half width, then the temperature dependence of γ cancels that of β , and the equivalent width for a band of strong non-overlapping pressure broadened lines is independent of temperature.

Numerical values of the equivalent width of the $15\ \mu\text{m}$ band are calculated from (A.7) and the band strengths and isotopic abundances listed in Table A.1. The strength of the band is given by

$$\alpha = \left(\sum_{i,n} \sqrt{q_i r_{in} \alpha_n} \right) \quad (\text{A.8})$$

where q_i is the abundance of the i th isotope; r_{in} is the "line present" parameter listed in the table; and α_n is the strength of the n th band. The half-widths of the lines broadened by N_2 at 300°K is taken to be $0.71\ \text{cm}^{-1}\text{atm}^{-1}$ (Yamamoto,). With these values, the equivalent width of non-overlapping lines is given by

$$A_0 = 1901 \left(1 - e^{-960/U} \right)^{3/2} \cdot \left(1 + 3.41 e^{-480/U} + 7.04 e^{-960/U} \right) \left(\frac{P}{P_0} U \right)^{1/2} \quad (\text{A.9})$$

where A is in cm^{-1} and U is in gm cm^{-2} . The exponentials in (A.9) allow for the population of the first and second vibrational levels at higher temperatures and for stimulated emission.

When the lines overlap each other, the equivalent width of the band is taken to be

$$A = C \ln (1 + A_0/C)$$

In (A.10) C is derived by fitting the expression to laboratory measurements. From the measurements obtained by Burch et al. (1962) we obtain $C = 48 (T/T_0)^{1/2} \text{cm}^{-1}$.

The temperature dependence assigned to C is that for the equivalent width of the band in the limit of completely overlapped lines (Cess and Tiwari,). It arises because at higher temperatures, rotational levels with large J become populated and the width of the band grows. Laboratory measurements of the equivalent width, however, are usually taken at room

temperatures or higher, and at such temperatures the hot bands contribute significantly to the absorption. The measurements from which C was derived were taken near 300°K, and therefore, the value derived for C contains contributions from the hot bands. At low temperatures the hot bands are not present and the value of C obtained from laboratory measurements at low temperatures could differ from that obtained with the temperature dependence assigned above.

TABLE A-1. ABUNDANCE OF CO₂ ISOTOPES AND STRENGTHS OF BANDS IN 15 μ m REGION (DRAYSON, 1967). "Lines Present" indicates whether every other line is missing, 1, or present, 2.

| <u>Isotope</u> | <u>Abundance</u> | <u>Lines Present</u> |
|--|------------------------|----------------------|
| C ¹² O ¹⁶ | 0.987 | 1 |
| C ¹³ O ¹⁶ | 1.1 x 10 ⁻² | 1 |
| C ¹² O ¹⁶ ₂ ¹⁸ | 3.9 x 10 ⁻³ | 2 |
| C ¹² O ¹⁶ ₂ ¹⁷ | 7.9 x 10 ⁻⁴ | 2 |
| C ¹³ O ¹⁶ ₂ ¹⁸ | 4.4 x 10 ⁻⁵ | 2 |

| <u>Band</u> | <u>Transition</u> | <u>Strength (300°K)</u> <u>cm⁻¹ (atm-cm)⁻¹</u> | <u>Lines Present</u> |
|-------------|-------------------------------------|---|----------------------|
| 1 | 00 ⁰ 0-01 ¹ 0 | 194 | 1 |
| 2 | 01 ¹ -02 ⁰ 0 | 4.27 | 1 |
| 3 | 01 ¹ 0-10 ⁰ 0 | 5.0 | 1 |
| 4 | 01 ¹ 0-02 ² 0 | 15.0 | 2 |
| 5 | 02 ⁰ 0-03 ¹ 0 | 1.0 | 1 |
| 6 | 02 ⁰ 0-11 ¹ 0 | 0.022 | 1 |
| 7 | 02 ² 0-11 ¹ 0 | 0.12 | 2 |
| 8 | 02 ¹ 0-03 ¹ 0 | 0.14 | 2 |
| 9 | 02 ² 0-03 ³ 0 | 0.85 | 2 |
| 10 | 10 ⁰ 0-03 ¹ 0 | 0.004 | 1 |
| 11 | 03 ³ 0-04 ² 0 | 0.0042 | 2 |
| 12 | 03 ³ 0-12 ² 0 | 0.0059 | 2 |
| 13 | 03 ¹ 0-12 ² 0 | 0.00049 | 2 |
| 14 | 03 ¹ 0-12 ⁰ 0 | 0.014 | 1 |
| 15 | 10 ⁰ 0-11 ¹ 0 | 0.3 | 1 |

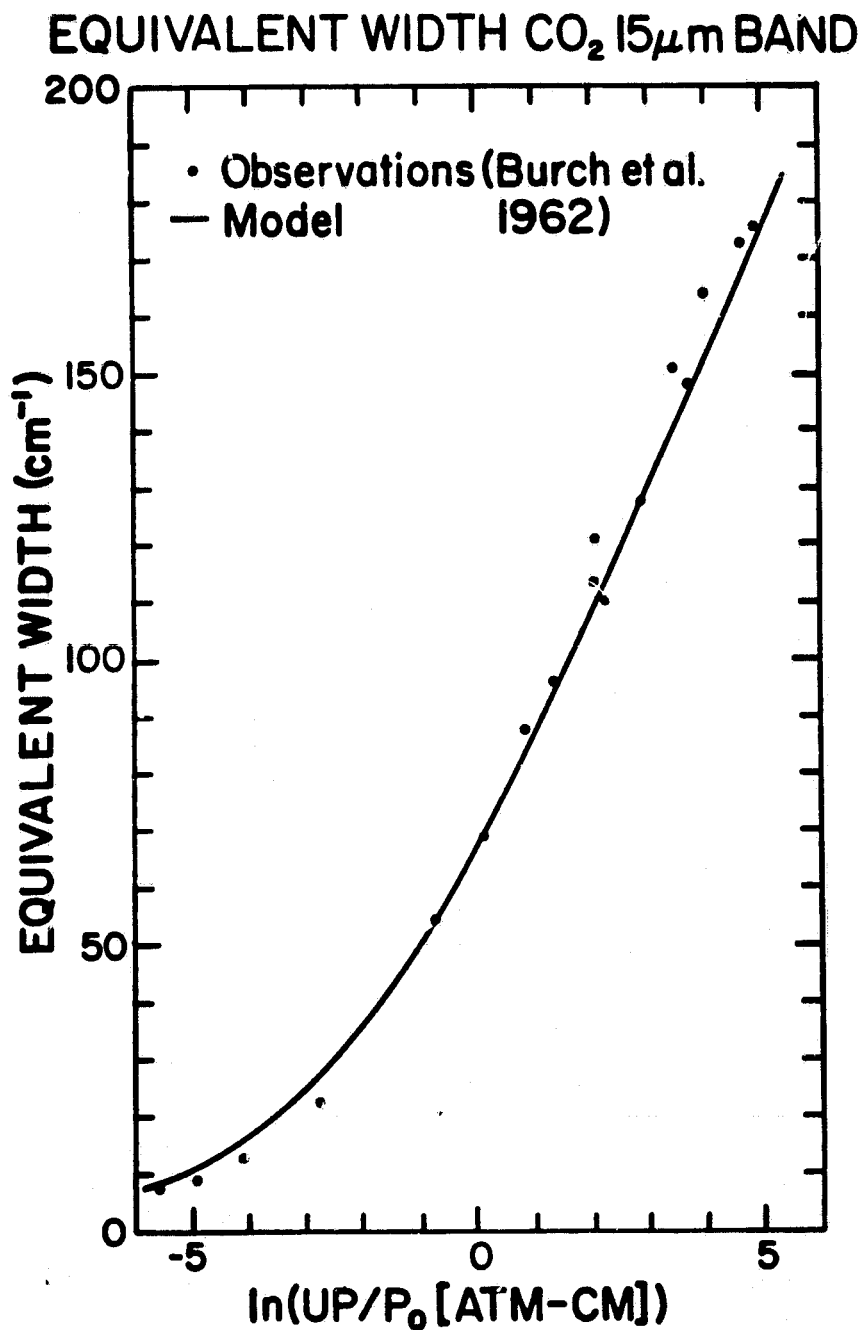


FIGURE A-1. EQUIVALENT WIDTHS MEASURED BY BURCH ET AL. (1962) (DOTS) AND COMPUTED FROM TABLE A-1 (SOLID LINE). The measurements shown in the figure have pressure broadened path lengths equivalent to the path-to-space pathlength in the earth's atmosphere with CO₂ concentrations ranging from 1/2 to 10 times the present concentration of 330 ppmv.

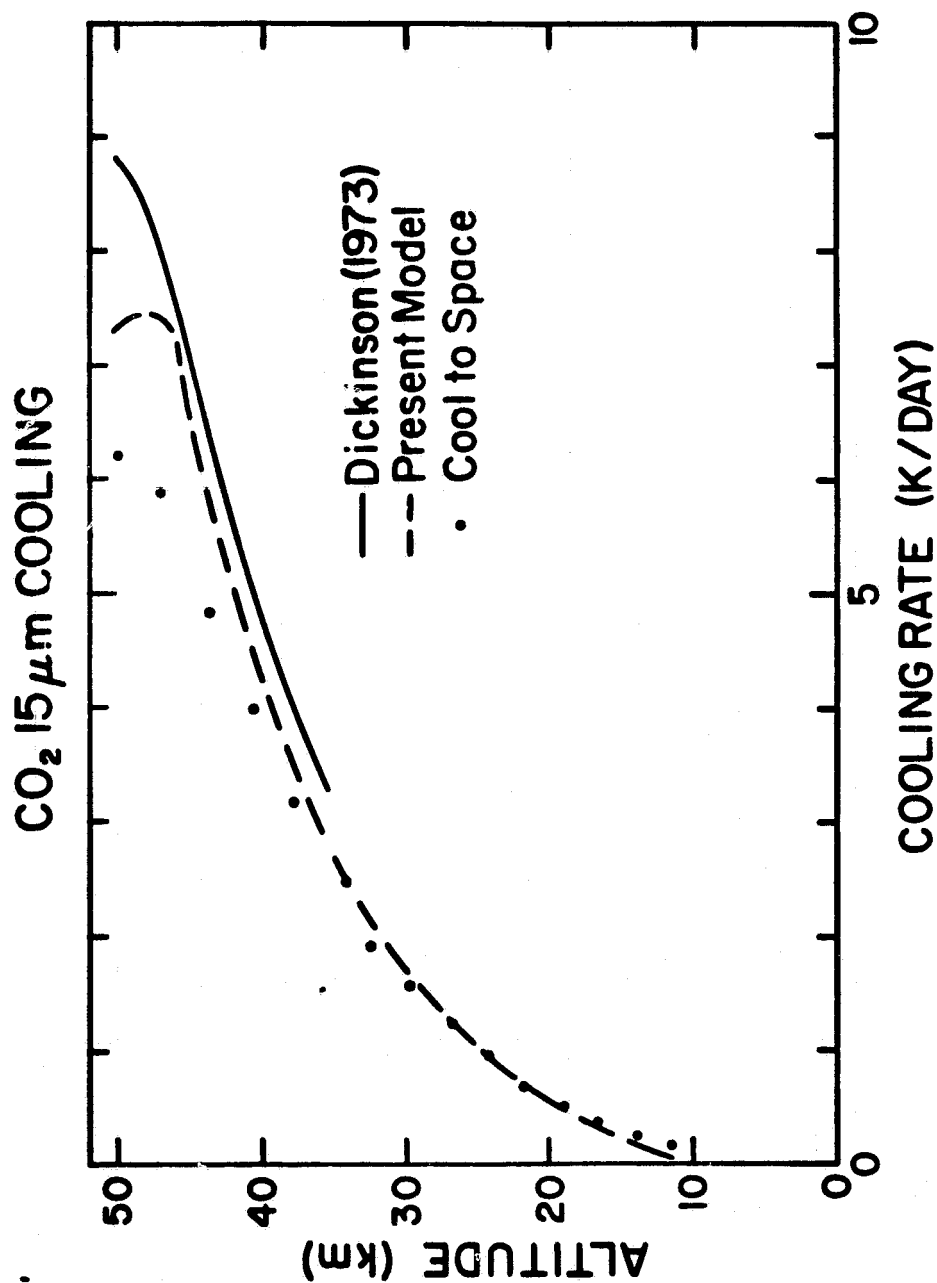


FIGURE A-2. COOLING RATES CALCULATED BY DICKINSON (1973) (SOLID CURVE) AND OBTAINED FROM TABLE A-1 (DASHED CURVE). The cool-to-space contribution to the cooling rates is shown as dots and indicates the importance of the path-to-space path-length for the cooling of the stratosphere.

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